PREPRINT UCRL- 81780

Lawrence Livermore Laboratory

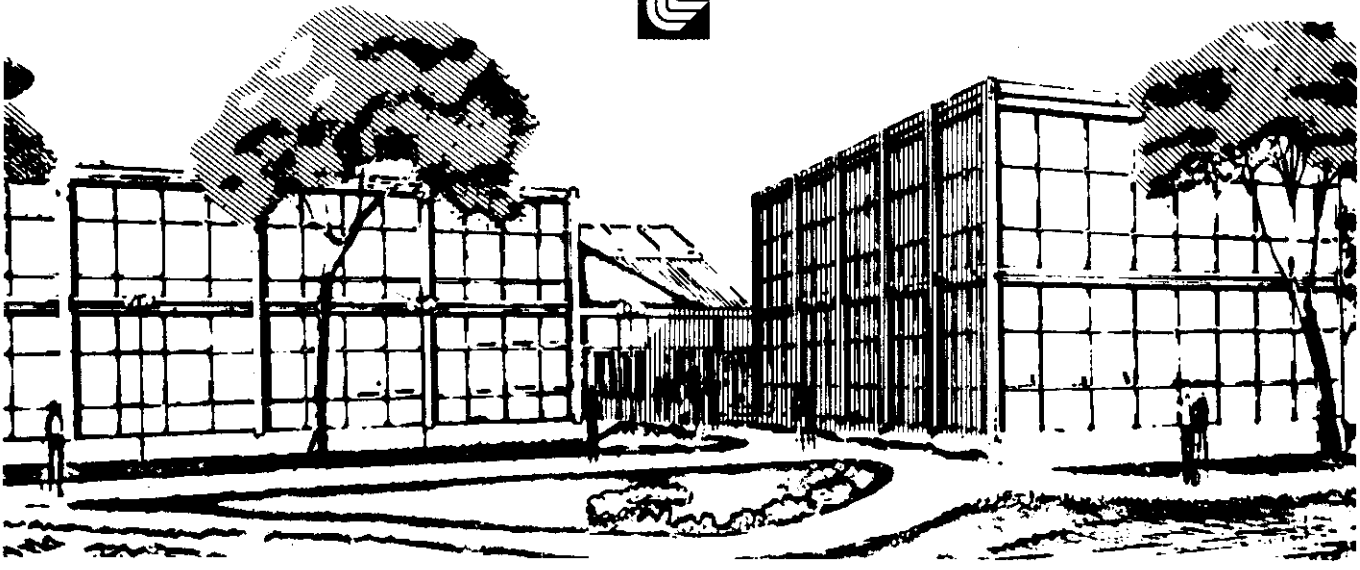
A REVIEW OF MEASUREMENT TECHNIQUES FOR STACK MONITORING OF LONG-LIVED ALPHA EMITTERS

Joseph F. Kordas and Paul L. Phelps

Electronics Engineering Department and Environmental Sciences Division

November 1, 1978

This paper was prepared for presentation at the 26th IEEE Nuclear Science Symposium and publication in the IEEE Transactions on Nuclear Science, Washington, D.C., October 18-20, 1978.



A REVIEW OF MEASUREMENT TECHNIQUES FOR STACK MONITORING OF LONG-LIVED ALPHA EMITTERS

Joseph F. Kordas and Paul L. Phelps*

Summary

As a result of the promulgation of new guidelines by the Environmental Protection Agency (40 CFR 190) for releases of long-lived, alpha-emitting substances, the stack-monitoring requirements for measuring long-lived alpha particles may change in terms of both monitored isotopes and the detection levels. This paper briefly reviews stack-monitoring requirements for long-lived alpha-emitting particles. It also examines the currently deployed alpha-particulate, stack-monitoring systems and discusses prototype systems that may be applicable to stack monitoring.

Introduction

Nuclear laboratories, processing facilities, and fabrication plants handle large quantities of long-lived, alpha-emitting substances. Large, sophisticated air-cleaning systems and highly sensitive monitoring systems are necessary to ensure that as little as possible of these materials enter the environment. In the past, most effort was centered on monitoring plutonium and uranium in stack effluents. However, the promulgation of new guidelines by the Environmental Protection Agency (40 CFR 190) for releases of long-lived alpha emitters from the nuclear fuel cycle may require monitoring the release of other isotopes including other alpha-emitting transuranics from processing facilities and ^{230}Th and ^{226}Ra from uranium mills.[1] These same regulations may also increase the demand for measurement systems with higher sensitivity. Some of the elements of interest along with their alpha energies, half-lives, and 40-h occupational maximum permissible concentrations (MPC) are listed in Table 1. Note that the MPC of ^{230}Th is almost as restrictive as that of plutonium.

Table 1. Some long-lived, alpha-emitting isotopes present in stack effluents of the nuclear fuel-cycle facilities.

Element	Half-life (y)	Alpha energy (MeV)	Maximum permissible concentration in air (pCi/l)
^{226}Ra	1,600	4.78, 4.60	0.03
^{230}Th	80,000	4.68, 4.62	0.002
^{232}U	73.6	5.32, 5.26	0.03
^{238}Pu	86	5.50, 5.46	0.002
^{239}Pu	24,400	5.15, 5.13, 5.10	0.002
^{240}Pu	6,580	5.17, 5.12	0.002
^{241}Am	458	5.48, 5.44, 5.39	0.006
^{243}Am	7,950	5.27, 5.22, 5.17	0.006
^{242}Cm	0.45	6.11, 6.07	0.1
^{244}Cm	18.1	5.80, 5.76	0.009

Currently, alpha detection is the only practical method available to measure low-level transuranic emissions.[2,3] This also may apply to ^{230}Th. Gamma intensities of the transuranics are extremely low except for the 59.6-keV gamma of ^{241}Am, 0.359 gamma/decay.[4] However, x-ray measurement seems more promising. The intensities are higher (0.0465 xrays/alpha for ^{239}Pu), but the spectra are complex and difficult to interpret.[4] Surface-ionization mass spectrometry may be a useful technique to monitor low-level emissions.[5,6] However, it is relatively new and untried and very complex for multi-isotope measurement.

This paper examines the currently deployed alpha-particulate, stack-monitoring systems and discusses prototype systems that may be applicable to stack monitoring. Much of the information in this paper is discussed in greater depth in Ref. 7.

Monitoring Requirements

Environmental Restrictions

An online-monitoring system for particles containing long-lived alpha emitters in stack effluents in many cases must be able to withstand the corrosive effluent stream (high moisture content and high acidity) and yet detect small quantities of long-lived alpha emitters in the presence of a much larger natural alpha background.

The daughters of ^{222}Rn (radon) and ^{220}Rn (thoron) constitute natural alpha background for long-lived, alpha-particulate measurement; ^{222}Rn and ^{220}Rn are gases, but their daughters are charged and readily attach to particles that are collected with the long-lived alpha emitters.[8] In 1952, Wilkening found that most of the natural alpha activity in air resulting from radon, thoron daughters is associated with particles with diameter <0.04 μm.[9] Under normal conditions, ^{222}Rn and ^{220}Rn concentrations 1 m above ground level range from 0.04 to 0.4 pCi/l.[10] This is 20 to 200 times the 40-h occupational MPC of ^{239}Pu. The decay schemes of ^{222}Rn and ^{220}Rn are given in Fig. 1. Figure 2 illustrates the alpha spectrum of their daughters.

The interfering daughters are ^{218}Po (RaA) and ^{212}Bi (ThC), emitting alphas at 5.99 and 6.05 MeV, respectively. Also, ^{214}Po (RaC') emits an alpha at 7.68 MeV that interferes to a lesser degree. The long interfering tail of the spectrum of the 7.68-MeV alpha of ^{214}Po results more from alpha-energy degradation caused by penetration of the filter paper than from degradation resulting from air.

Effluent streams from processing plants and scrap-recovery plants are extremely corrosive. Sensitive, solid-state detectors cannot withstand direct contact with these corrosive streams. Therefore, a less sensitive, but more corrosive-resistant, detection system may have to be employed at these facilities.

*Electronic Engineering Department and Environmental Sciences Division, Mail Stop L-524, Lawrence Livermore Laboratory, P. O. Box 5507, Livermore, CA 94550

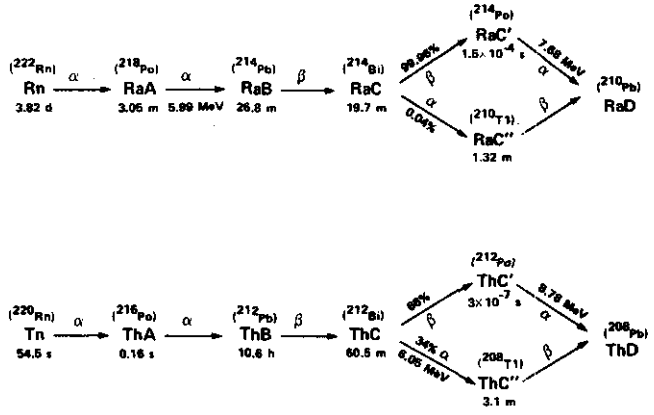


Fig. 1 Decay schemes of radon, ^{222}Rn (above), and thoron, ^{220}Rn (below).

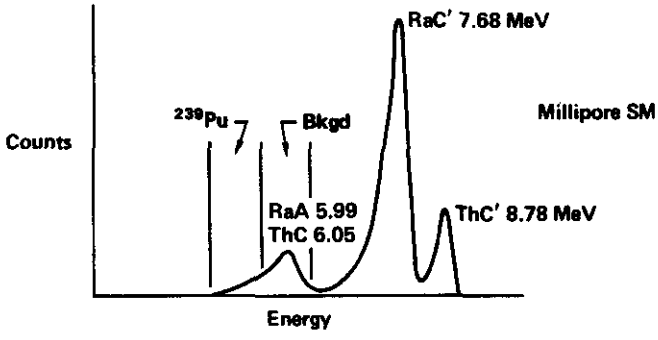


Fig. 2 Alpha spectrum of natural background counted on a Millipore SM filter at 1-atm pressure.

Detection Vs Measurement

"Following an experimental observation, one must decide whether or not that which was being sought was, in fact, detected. Formally known as Hypothesis Testing, such a binary, qualitative decision is subject to two kinds of errors: deciding that the substance is present when it is not (α; error of the first kind) and the converse, failing to decide it is present when it is (β; error of the second kind).[11] Both of these kinds of errors must be considered when establishing a detection limit. In addition, the magnitude of these errors must be specified for the given detection limit.

The detection limit of commercial alpha-particulate monitors is specified usually in terms of the ANSI standard N 13.10 - 1974.[12] However, this standard assumes a constant background and also addresses only the error of the first kind. That is, when $n_s \geq 2\sqrt{n_B/2RC}$, where n_s is the signal count rate, n_B the background count rate, and RC the instrument time constant, we can be 95% confident that the substance of interest is present. This says nothing about how often it is present at this detection limit and goes undetected (error of the second kind). Here α = 5%, but β is unknown. Currie defines the detection limit as that level of activity for which the probability of an error of the first kind is α, and that of the second kind is β.[11] At the level where $\alpha = \beta = 0.05$, we can be 95% confident of detecting activity if present at the detection limit, and also 95% confident that when detection is indicated, the substance is truly present. Table 2 illustrates the difference in these two detection limits for a constant air monitor (CAM).

Table 2. Detection limits of a constant air monitor (CAM) after 30 h of operation.

Method [a]	Limit [a]
ANSI N 13.10-1974 $n_s = 2\,\sigma_o$	n_s = 15.7 cpm, or 5.8 h for detection of 1 MPC ^{239}Pu.
Currie $\alpha = \beta = 0.05$ $n_s = 2.71 + 3.29\,\sigma_o$	n_s = 28.4 cpm, or 10.6 h for detection of 1 MPC ^{239}Pu.
Fractional standard deviation $n_s = 3\,\sigma_s$ $n_s = \frac{9 + \sqrt{81 + 16\,n_B}}{2}$	n_s = 28.4 cpm, or 10.6 h for detection of 1 MPC ^{239}Pu.

[a] Where n_s and σ_s are the true signal count rate and its standard deviation, n_B is the count rate in the background window, and σ_o is the standard deviation of the true signal when n_s = 0. MPC is the 40-h occupational MPC (0.002 pCi/l).

In many instances, neither a binary decision nor an upper limit of detection is satisfactory. A more quantitative measure of the activity is required. In this case, the sensitivity of the measurement technique is better represented by a determination limit at which a given procedure will be sufficiently precise to yield a satisfactorily quantitative estimate.[11] Table 2 includes the determination limit of a CAM measurement with a fractional standard deviation of 0.33, assuming the same conditions as the detection limits.

Measurement of Routine Vs Accidental Releases

The system requirements for online monitoring of routine releases of long-lived alpha emitters in stack effluents are different than those for monitoring accidental releases of the same isotopes. In the first case, to detect malfunctions in the air-cleaning system before serious releases occur and to assure compliance with release guidelines, the online system must be able to measure routine emissions quantitatively. This requires extremely high sensitivity on almost a real-time basis. In the second case, the online system must respond immediately to a high level release and be able to follow it to extremely high levels. This requires a system with rapid response and wide dynamic range.[13] However, its sensitivity could be considerably lower than that of the routine-release monitor. Whether these two sets of requirements can be met by the same system is uncertain.

Deployed Measurement Systems

Gross Alpha Measurement

On-line-monitoring systems based on gross alpha measurement do not discriminate against natural alpha background. Therefore, their sensitivity is poor compared to that of constant air monitors. However, they are more resistant to corrosive stack gas.

A gross-alpha monitoring system with a moving filter is pictured in Fig. 3.[14] It consists of a continuously moving filter (cellulose and glass-fiber) and two zinc sulfide detectors. A 56-lpm sample is routed through the moving filter (1.3 cm/h). The filter paper is scanned by the first zinc sulfide detector during collection (prompt channel) and by the second detector 8 h after collection (delay channel). In both cases, gross alpha emission is counted.

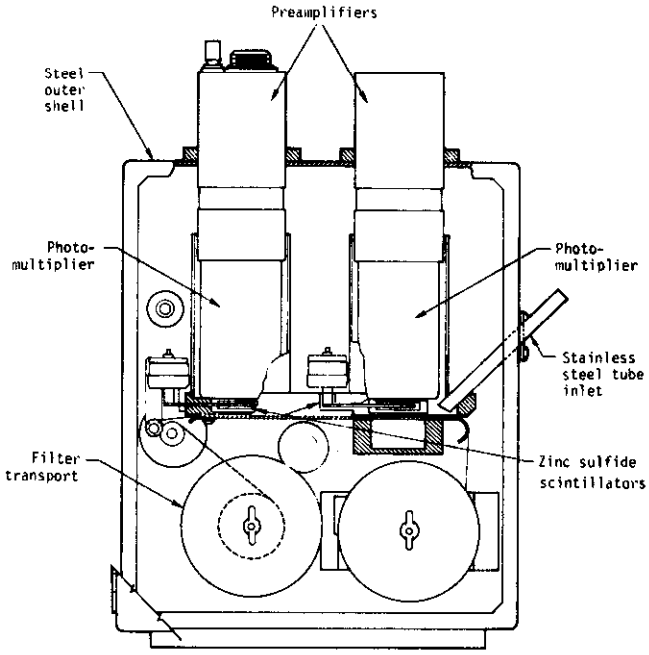


Fig. 3 A gross-alpha monitoring system with moving filter paper.[14]

The limitations of this type of monitor are obvious, the most important is its extremely low sensitivity. This is reflected in the proposed alarm level of the prompt channel, 10 times background or 1500 MPC-h of ^{239}Pu for an alpha background of 890 cpm.[7,14] The instrument reaches equilibrium in 1 h at constant activity. Therefore, the equivalent of 1500 MPC of ^{239}Pu would be required to alarm the instrument. This type of instrument also would not be able to distinguish the long-lived alpha emitters from some of the shorter-lived alpha emitters such as ^{242}Cm present in some effluent streams.

Constant Air Monitor (CAM)

The constant air monitor (CAM) is the most popular online-detection system for long-lived alpha emitters. It used both alpha spectroscopy and a background-compensation scheme to reduce interference from natural background.

The sampling head of a CAM is illustrated in Fig. 4. A 40 to 60 lpm sample is drawn through a fixed membrane filter. A solid state detector with a diffused junction scans the filter. The detector feeds two single-channel analyzers, one for ^{239}Pu (4.8 to 5.2 MeV) or ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Am (4.8 to 5.5 MeV) and the other for the natural background ^{218}Po and ^{212}Bi (5.6 to 6.2 MeV). This is depicted in Fig. 2. The natural background is discriminated against by alpha spectroscopy. However, the energy degradation of the alphas by the filter paper and air allows approximately 25% of the natural background resulting from ^{218}Po and ^{212}Bi to enter the ^{239}Pu window.[15] To compensate for the 25%, a preset percentage of the activity

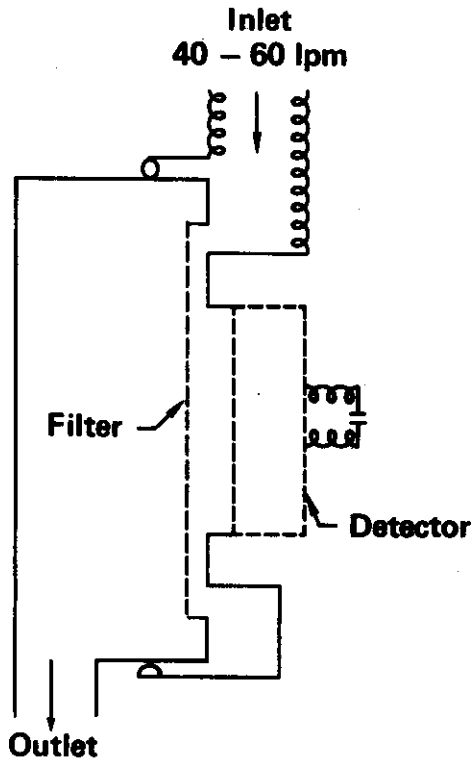


Fig. 4 The sampling head of a constant air monitor (CAM).

entering the background window (5.6 to 6.2 MeV) is subtracted from the activity entering the Pu window. One problem with this type of background compensation is that the percentage of background activity entering the Pu window can change. It depends on both the particle size of background particles and filter loading.

The detection limit of a CAM after 30 h of operation is given in Table 2. These calculated limits assume background concentrations of 0.1 and 0.01 pCi/l for ^{218}Po and ^{212}Pb, respectively, in the sampled air, a detector efficiency of 17% of 4π, and a time constant of 30 s. A factor of 25% was assumed for the background resulting from ^{218}Po and ^{212}Bi entering the ^{239}Pu window. No variations in this factor were considered. Note the difference in the detection limits as defined by the ANSI standard and by Currie, $\alpha = \beta = 0.05$. Under these conditions, the CAM must sample ^{239}Pu at 1 MPC (40-h occupational) for approximately 10 h before it can detect the activity at the 95% confidence level ($\alpha = \beta = 0.05$). The CAM's

sensitivity decreases for ^{238}Pu and ^{241}Am because a larger percentage of the natural alpha background enters the 4.8 to 5.5-MeV window. The sensitivity of the CAM is considerably better than that of the gross alpha-measurement system. However, the CAM is much more susceptible to failure because of direct contact of the sensitive solid-state detector with the effluent stream.

Prototype Measurement Systems

At least three prototype online-measurement systems for long-lived alpha emitters are under development: the Direct-Inlet Mass Spectrometer (DIMS) at Battelle Pacific Northwest Laboratory, the Virtual Impactor Solid-State Detector Online-Monitoring System at Argonne National Laboratory and the Transuranic Aerosol Measurement System (TAMS) at the Lawrence Livermore Laboratory. All three emphasize improved sensitivity.

Direct-Inlet Mass Spectrometer (DIMS)

Investigators at Battelle Laboratory are studying the feasibility of using direct-inlet, surface-ionization mass spectroscopy to analyze ^{239}Pu-containing particles in air. [7,16] This technique could yield not only the ^{239}Pu concentration in air, but also the distribution of sizes of particles containing ^{239}Pu. Figure 5 shows the particle path within the surface-ionization mass spectrometer.[17] The particle-laden air is pulled through a capillary nozzle at a rate of 5 cm^3/s. Inside the first vacuum chamber, the air expands and is pumped away, but the momentum of the particles carries them into a second vacuum chamber. Again, the residual air expands and is pumped away. The particles continue through a collimator and impinge on a rhenium filament at 1275°K and 10^{-3} Pa. The ions produced as the particles evaporate from the surface are withdrawn by an electric field, focused, and analyzed by a 15-cm-radius 60° magnet. The ions selected by the magnetic field impinge on an aluminum target held at -40 kV. The secondary electrons emitted from the target then pass into a plastic scintillator and the photons are counted.

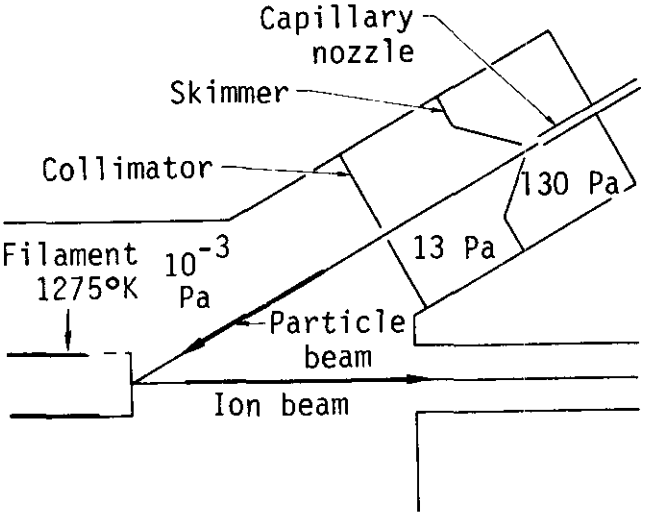


Fig. 5 The particle path within BNWL's surface-ionization mass spectrometer.[17]

At sufficiently high filament temperatures, each particle produces a short burst of ions. By counting the bursts, the number of particles/cm^3 of air can be measured. The number of ions per burst is a measure of the quantity of the element per particle. Assuming that the composition of particles is constant, this yields the distribution of sizes of particles. Theoretically, a surface-ionization mass spectrometric system could measure 10^3 to 10^4 atoms of ^{239}Pu, corresponding to particles of $^{239}PuO_2$ with diameters from 4.0 to 9.0 nm.

Many obstacles must be overcome before surface-ionization mass spectroscopy can be implemented as a stack-monitoring system. A summary of these follows.

- The DIMS system under development can only measure one isotope, ^{239}Pu. Expanding the present design to measure multi-isotopes simultaneously is not trivial.
- Because of ^{238}U interference, ^{238}Pu cannot be detected by surface-ionization mass spectroscopy.[5]
- Whether a surface-ionization mass spectrometer could withstand the corrosive onslaught of the stack effluent is questionable.
- The low inlet-flow rate drastically reduces instrument sensitivity to larger particles. Table 3 illustrates the problem.

Table 3. Relationship between particle-size distribution and average time to detect $^{239}PuO_2$.[a]

Physical Diameter μm	Aerodynamic diameter μm	Atoms/particle	Particles/l at 1.0 MPC	Average Particle detection time
0.1	0.34	1.3×10^7	6.1	33.0 s
0.5	1.7	1.7×10^9	0.049	1.1 h
1.0	3.4	1.3×10^{10}	0.0061	9.0 h

[a] A sampling rate of 5 cm/s is assumed.

Virtual-Impactor, Online-Monitoring System

The design of the Argonne virtual-impactor monitoring system for long-lived alpha-emitting particles is based on Wilkening's observation that most natural alpha activity is associated with very small particles. This system uses a combination of particle-size selection via a two-stage virtual impactor and alpha-energy spectroscopy to distinguish particles containing long-lived alpha activity from those containing only natural alpha activity.[18] An impactor-type plutonium-monitoring system has been deployed successfully as a hood monitor for many years at Argonne, the ZPR-9 Airborne-Plutonium Monitoring System.[19] It is conceptually similar to the virtual-impactor system except that it impacts the particles directly on the detector surface.

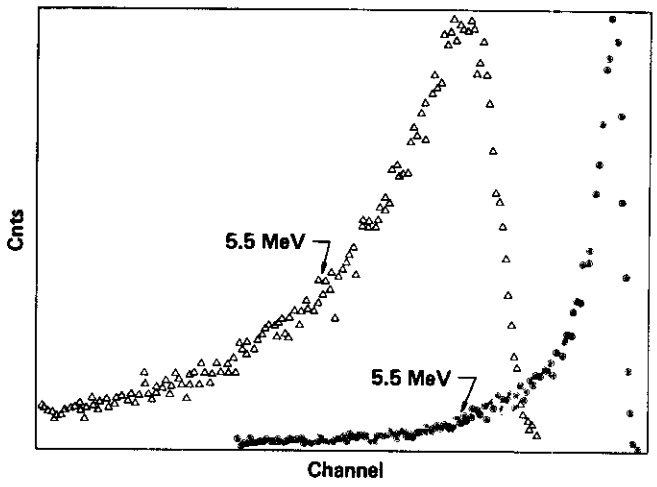


Fig. 7 Alpha spectra of ^{212}Bi (ThC - 6.05 MeV) acquired at 20 mm Hg and at 760 mm Hg after collecting natural background on Acropor filter paper for 24 h.

Figures 8 and 9 contain a block diagram of the TAMS system and a drawing of its filter-transport mechanism. A 566 slpm sample is drawn through a membrane filter paper (Acropor 1200) for a fixed period after which the filter containing the sampled particles is stepped in front of an array of four 600-mm^2, boron-implanted, surface-barrier detectors. Then the detector chamber is evacuated and the sample analyzed while another sample is collected. The entire process is controlled by an LSI-11 processor. The system requires approximately 22 s to vent the detector chamber, advance the filter paper, and pump the detector chamber back down to 20 mm Hg.

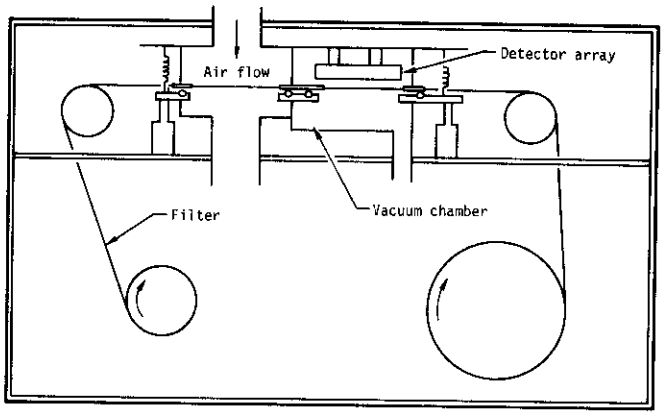


Fig. 9 Filter transport mechanism of the transuranic aerosol measurement system.

Figures 10 and 11 contain alpha spectra of radon, thoron background collected at Lawrence Livermore Laboratory by TAMS on Acropor 1200, and Fluoropore FA 1.0 filter paper, respectively. In both cases, the sample was collected and counted for 30 min. Note the resolution of the background peaks. In both cases, the center peak resulting from ^{214}Po (RaC' - 7.69 MeV) has a fwhm of about 80 keV. Also note that there is more tailing of the peaks with the AN 1200 filter. This tailing is related to the alpha-energy degradation resulting from filter paper penetration by small particles containing natural alpha background. Obviously, the small particles penetrate deeper into the Acropor 1200 than the Fluoropore FA 1.0 filter paper. In fact, an alpha spectrum of a plated ^{241}Am source on plutonium with a separation between source and detector of 5.0 mm is no better resolved than the background spectrum on Fluoropore filter paper. However, this filter paper is much more difficult to transport.

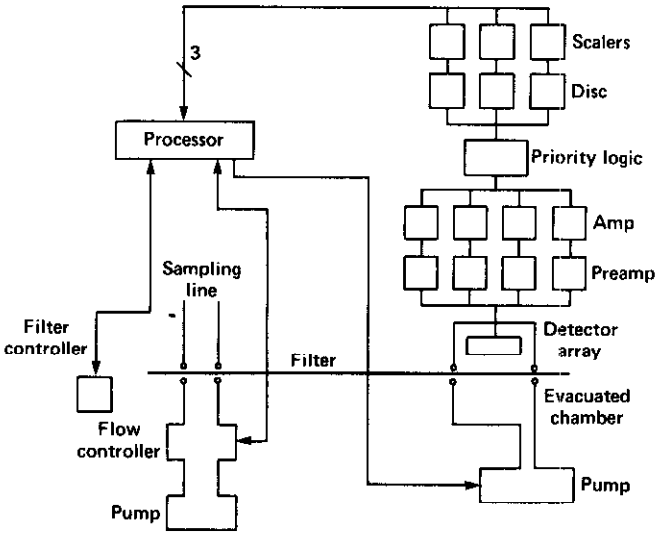


Fig. 8 The transuranic aerosol measurement system block diagram.

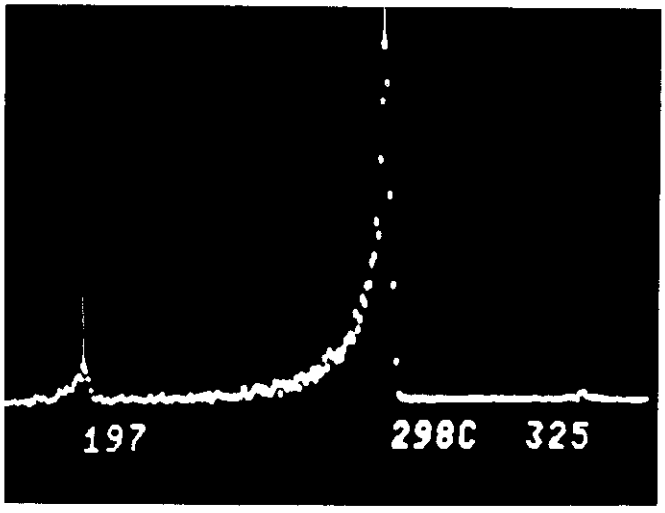


Fig. 10 Alpha spectrum of radon, thoron background collected by TAMS on Acropor 1200 filter paper.

The virtual impactor is illustrated in Fig. 6.[18] The first stage consists of nine virtual impactors with a total inlet flow of 283 lpm. The second stage uses only three virtual impactors. The large particles containing the long-lived alpha emitters pass through both impactor stages and are collected on the filter, while the smaller particles containing the natural alpha background follow the flow streamlines and are pumped away. Each impactor stage reduces the flow by 75%. Therefore, approximately 7% of the initial flow passes through the filter resulting in a filter collection of 7% of the small particles. This reduced flow results in a pressure drop of only 80 mm Hg with a Millipore AA filter. The cut point, the particle size at which the collection efficiency is 50%, corresponds to an aerodynamic diameter of 1.5 μm, or a real diameter of 0.4 μm, for $^{239}PuO_2$ particles.

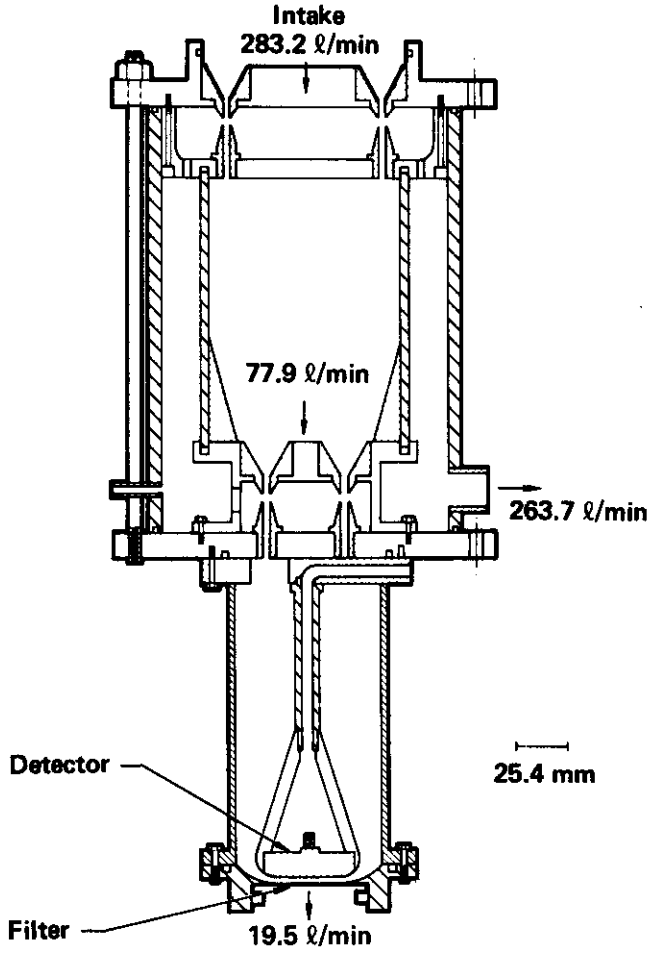


Fig. 6 Detailed drawing of the virtual-impactor solid-state detector online-monitoring system.[18]

An 800-mm^2 diffused-junction detector (30% of 4π efficiency) analyzes the material collected on the filter. A combination of alpha-energy spectroscopy and background compensation discriminates against the residual natural alpha activity on the filter. The counts in the selected energy windows are corrected for background by subtracting a fraction of the counts in an upper background window. The counts are summed for a fixed period of 15 to 60 min. The scalers then reset and the counting cycle begins over.

The attractive features of this system included:

- A high inlet flow rate of 280 lpm vs 60 to 100 lpm for the CAM.
- A high sensitivity capable of detecting 1 MPC_a (40-h occupational) in less than 1/2 h.
- A simultaneous collection and monitoring capability.

The low small-particle-collection efficiency and the probable susceptibility of an impactor system to the onslaught of the stack effluent may limit the applicability of the virtual-impactor system to stack monitoring. Its particle-size cutoff occurs at an aerodynamic diameter of 1.5 μm. Yet, Elder et al.[20] found that a large fraction of the PuO_2 in the ventilation system at a plutonium recovery plant consisted of particles with aerodynamic diameters < 1.0 μm. The HEPA filters (high-efficiency particulate air filter) would concentrate further the particle-size distribution into the 0.2 to 0.6 μm region.[21]

Transuranic Aerosol Measurement System

The Transuranic Aerosol Measurement System (TAMS) combines a high sampling rate with high-resolution alpha spectroscopy and decay analysis to achieve extremely high sensitivity.[7,22] It was designed specifically to measure low concentrations of long-lived alpha particles in harsh atmospheres such as corrosive stack effluents. Some of the more prominent features of this measurement system are:

- Separate collection and counting chambers to completely isolate detectors from the effluent stream,
- An evacuated detection chamber that improves the spectral resolution by more than five-fold,
- A high inlet flow rate, 566 slpm,
- Decay scheme analysis to eliminate residual, natural alpha background resulting from ^{218}Po based on the difference in lifetimes of the long-lived alpha emitters (y) and that of ^{218}Po (min).

A disadvantage of this type of measurement system is that collection and detection are no longer simultaneous. Therefore, a delay equivalent to the collection time could exist between a high level release and its detection.

Figure 7 illustrates the drastic improvement in spectral energy resolution obtained by evacuating the detection chamber. The full width at half maximum (fwhm) of the spectra taken at 20 and 760 mm Hg are 110 and 572 keV, respectively. The percentage of activity below 5.5 MeV in the two spectra are 6.0 and 25%, respectively. Any residual background resulting from ^{212}Bi after alpha spectroscopy is compensated by subtracting a fraction of the activity in an upper window (^{212}Po window, 8.78 MeV). Because of the short half-life of ^{212}Po (3×10^{-7}s), ^{212}Po is always in secular equilibrium with ^{212}Bi.

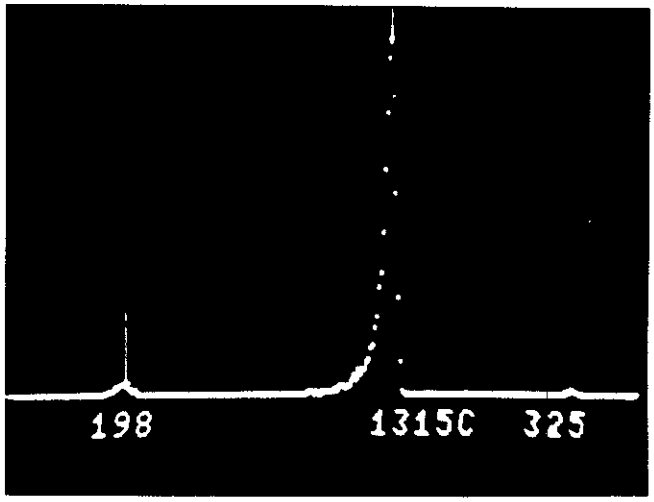


Fig. 11 Alpha spectrum of radon, thoron background collected by TAMS on Fluoropore FA 1.0 filter paper.

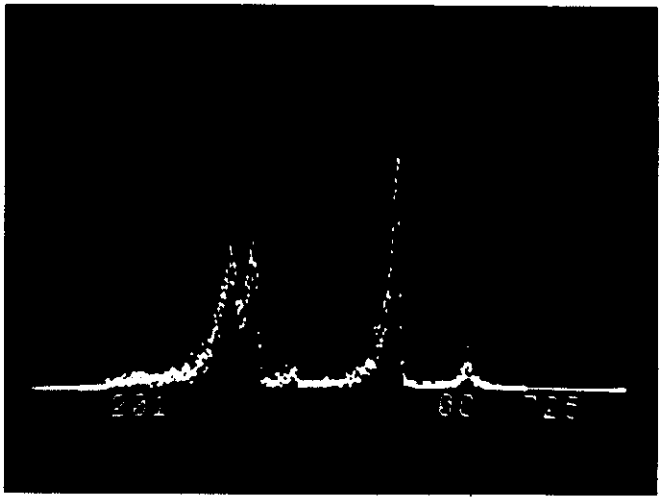


Fig. 12 Alpha spectrum of a stack sample collected for 60 min by TAMS at a plutonium recovery facility.

Figure 12 contains an alpha spectrum of a stack sample collected by TAMS during field testing at a plutonium-recovery facility. This spectrum illustrates the spectral resolution of the TAMS system. This is a spectrum of particles collected on an Acropor 1200 filter for 60 min and counted for 50 min. Note the high-resolution of the background peaks. The HEPA filters very efficiently remove the small background particles that caused the tailing noticeable in Fig. 10. The two peaks on the left are a result of ^{239}Pu (5.15 MeV) and ^{241}Am (5.48 MeV). Note that TAMS can resolve these peaks. This spectrum of ^{239}Pu and ^{241}Am represents a combined release of approximately 5.0 MPC_a. However, this spectrum does not actually result from a release, but a sampling artifact caused by filter contamination from condensation in the sampling lines.

The results of field testing TAMS indicate that the natural background is so well resolved that decay analysis is unnecessary and that the TAMS system is only limited by counting statistics. Table 4 lists fractional standard deviations of TAMS measurements with variations in long-lived alpha concentration in MPC_a (40-h occupational) and collection time. According to this table, TAMS can measure 1/40 of an MPC_a of ^{239}Pu in 60 min with an fsd of 0.18. This is true only for particles containing isotopes of low specific activity like ^{239}Pu. TAMS like DIMS has difficulty collecting large particles of isotopes with high specific activity like ^{238}Pu. This is illustrated in Table 5. If $^{238}PuO_2$ is released at 1.0 MPC_a (40-h occupational) in particles with diameters of 1.0 μm, TAMS would require 84 min on the average to collect one particle sampling at 566 slpm. We believe that this particle-collection limitation will limit the ultimate sensitivity level of measuring activity in particles containing high specific activity isotopes not only by TAMS but by any online-monitoring system.

Table 4. Fractional standard deviation for TAMS measurements with variations in concentration and collection time assuming a sampling rate of 566 slpm, collection efficiency of 100% and detection efficiency of 13.2% of 4π (MPC_a - 0.002 pCi/l).

Concentration (MPC_a)	Collection time (min)	Fractional standard deviation
1.0	15	0.12
0.25	15	0.23
0.1	30	0.18
0.1	60	0.092
0.025	60	0.18

Table 5. The dependency of collection time on particle size for PuO_2, assuming a collection efficiency of 100% and a sampling rate of 566 slpm.

Isotope	Half-life (y)	Physical diameter (m)	Aerodynamic diameter (m)	Particles/l at 1.0 MPC_a	Average collection time of 1 particle (min)
^{238}Pu	86	0.1	0.34	2.2×10^{-2}	8.0×10^{-2}
		0.5	1.7	1.7×10^{-4}	10.4
		1.0	3.4	2.2×10^{-5}	84.0
^{239}Pu	24,400	0.1	0.34	6.1	2.9×10^{-4}
		0.5	1.7	4.9×10^{-2}	3.6×10^{-2}
		1.0	3.4	6.1×10^{-3}	0.29

Acknowledgements

The authors would like to thank Lester Bruns of the Rockwell Hanford Operation, Kenneth Lamson of Lawrence Livermore Laboratory, James Stoffels of Battelle Pacific Northwest Laboratory and Thomas Yule of Argonne National Laboratory. Special thanks go to *Hodge Wasson, formerly of the Department of Energy,* Washington, D.C., for his foresight and encouragement in recognizing the value and need of this work.

References

1. Environmental Protection Agency, "Environmental Radiation Protection Standards for Nuclear Power Operations," 40 CFR 190, Federal Register Vol. 42, No. 9 (January 13, 1977).

2. R. J. Budnitz, "Plutonium: A Review of Measurement Techniques for Environmental Monitoring," IEEE Trans. Nucl. Sci., NS-21, 430 (1974).

3. "Alpha Particle Instrumentation" in Instrumentation for Environmental Monitoring, Lawrence Berkeley Laboratory, Rept. LBL-1, Vol. 3 (1973).

4. K. L. Swinth, "Photon Intensities and Importance in Counting Transuranic Materials," Battelle Pacific Northwest Laboratories, Rept. BNWL-1648 (1972).

5. C. E. Pietri, "The Determination of Plutonic Isotopic Composition by Mass Spectroscopy, and Americium-241 Content by Radiocounting," in Proc. Symp. Calorimetric Assay of Plutonium, W. W. Strohm and M. F. Hawenstein, Eds., Mound Laboratory, Monsanto Research Corporation, Rept. MLM-2177 (1973).

6. W. E. Davis, "Continuous Mass Spectrometric Analysis of Particulates and Other Impurities in Air and Water Using Surface Ionization," General Electric Corporation, Schenectady, New York (1975).

7. J. F. Kordas and P. L. Phelps, A Review of Monitoring Instruments for Transuranics in Fuel Fabrication and Reprocessing Plants," A Progress Report to the Physical and Technological Programs, Division of Biomedical and Environmental Research, U. S. Energy Research and Development Administration, Lawrence Livermore Laboratory, Rept. UCRL-52123, Rev. 1 (1977).

8. R. J. Budnitz, "Radon-222 and Its Daughters: A Review of Instrumentation for Occupational and Environmental Monitoring," Health Phys., 26, 145 (1974).

9. M. H. Wilkening, "Natural Radioactivity as a Tracer in the Sorting of Aerosols According to Mobility," Rev. Sci. Instrum. 23, 13 (1952).

10. Ionizing Radiation: Levels and Effects, (United Nations, New York 1972), Vol. 1, pp. 32.

11. L. A. Currie, "Limits for Qualitative Detection and Quantitative Determination: Applications to Radiochemistry," Anal. Chem., 40, 586 (1968).

12. Specification and Performance of On-Site Instrumentation for Continuously Monitoring Radioactivity in Effluents, American National Standards Institute, Inc., Rept. ANSI N13.10 (1974).

13. Technological Considerations in Emergency Instrumentation Preparedness, Phase II-B, Emergency Radiological and Meteorological Instrumentation for Mixed-Oxide Fuel Fabrication Facilities, Battelle Pacific Northwest Laboratories, Rept. BNWL-1742 (1974).

14. Final Safety Analysis Report, Barnwell Nuclear Fuel Plant License Application, Allied-Gulf Nuclear Services, Docket 50332-40 (1975).

15. W. A. Phillips and C. L. Lindeken, "Plutonium Alpha Air Monitor Using a Solid-State Detector," Health Phys., 9, 299 (1963).

16. J. J. Stoffels, P. J. Hof, and T. P. Harrington, "Direct-Inlet Mass Spectrometer Development," in Battelle Pacific Northwest Laboratory Annual Report - 1977, PNL-2500 Pt-4 (1978).

17. J. J. Stoffels, private communication (1976).

18. T. J. Yule, "An On-line Monitor for Alpha-Emitting Aerosols," IEEE Trans. Nucl. Sci., NS-25, 762 (1978).

19. G. K. Rusch and W. P. McDowell, "The ZPR-9 Airborne Plutonium Monitoring System," IEEE Trans. Nucl. Sci., NS-23, 690 (1976).

20. J. C. Elder, M. Gonzales and H. J. Ettinger, "Plutonium Aerosol Size Characteristics," Health Phys., 27, 45 (1974).

21. W. B. Seefeldt, W. J. Mecham, and M. J. Steindler, Characterization of Particulate Plutonium Released in Fuel Cycle Operations, Argonne National Laboratory, ANL-75-78 (1976).

22. J. F. Kordas and P. L. Phelps, "An Online High Sensitivity System for Transuranic Aerosols," IEEE Trans. Nucl. Sci., NS-24, 670 (1977).

"Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore Laboratory under contract number W-7405-ENG-48."